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Review

Real-time air quality forecasting, part II: State of the science, current research needs, and future prospects

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ABSTRACT

The review of major 3-D global and regional real-time air quality forecasting (RT-AQF) models in Part I identifies several areas of improvement in meteorological forecasts, chemical inputs, and model treatments of atmospheric physical, dynamic, and chemical processes. Part II highlights several recent scientific advances in some of these areas that can be incorporated into RT-AQF models to address model deficiencies and improve forecast accuracies. Current major numerical, statistical, and computational techniques to improve forecasting skills are assessed. These include bias adjustment techniques to correct biases in forecast products, chemical data assimilation techniques for improving chemical initial and boundary conditions as well as emissions, and ensemble forecasting approaches to quantify the uncertainties of the forecasts. Several case applications of current 3-D RT-AQF models with the state-of-the-science model treatments, a detailed urban process module, and an advanced combined ensemble/data assimilation technique are presented to illustrate current model skills and capabilities. Major technical challenges and research priorities are provided. A new generation of comprehensive RT-AQF model systems, to emerge in the coming decades, will be based on state-of-the-science 3-D RT-AQF models, supplemented with efficient data assimilation techniques and sophisticated statistical models, and supported with modern numerical/computational technologies and a suite of real-time observational data from all platforms.

1. Introduction

Part I of this review identified several inaccuracies in RT-AQF and their possible causes. Factors related to meteorology including inaccurate characterization of the transport (e.g., Eder et al., 2006; Yu et al., 2007, 2008) and planetary boundary layer (PBL) meteorological processes such as turbulent mechanisms and vertical convection, cloud attenuation of photolysis (e.g., Eder et al., 2006), local drainage and sea-breeze circulations (e.g., Hess et al., 2004; Honoré et al., 2008) and variables such as temperature, water vapor, inversion, and PBL heights (e.g., Berge et al., 2002; Hess et al., 2004; McKeen et al., 2007; Hogrefe et al., 2007). Factors related to boundary conditions (BCONs) include inadequate representations of BCONs of O_3, $PM_{2.5}$, and PM_{10} (e.g., McKeen et al., 2005; Yu et al., 2007; Chen et al., 2008; Chuang et al., 2011). Factors related to emissions include uncertainties in anthropogenic emissions of SO_2 (e.g., McKeen et al., 2007), NO_x (e.g., McKeen et al., 2005, 2009), and VOCs (e.g., Shrivastava et al., 2010), NH_3 (e.g., McKeen et al., 2007; Yu et al., 2008; Chuang et al., 2011), biogenic VOC emissions (e.g., McKeen et al., 2005, 2007; Yu et al., 2008; Hu et al., 2008; Chuang et al., 2011), wildfire emissions (e.g., Snow et al., 2003; McKeen et al., 2007; Chen et al., 2008), primary PM (e.g., Berge et al., 2002; Manins et al., 2002; McKeen et al., 2007; Hogrefe et al., 2007; Manders et al., 2009; Shrivastava et al., 2010; Chuang et al., 2011), dust (e.g., Jiménez-Guerrero et al., 2008), and pollen emissions (e.g., Sofiev et al., 2006). Factors related to model process treatments include inaccurate model treatments such as urban processes (Baklanov et al., 2002), gas-phase chemistry (e.g., Chen et al., 2008; Cai et al., 2008), in-cloud oxidation of SO_2 (Yu et al., 2008; Cai et al., 2008), SOA formation (McKeen et al., 2009; Shrivastava et al., 2010; Chuang et al., 2011), dry and wet deposition (McKeen et al., 2007; Hogrefe et al., 2007). A factor related to model configuration is the use of a coarse grid resolution (e.g., Cope et al.,

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2004). These studies indicate needs in improving several aspects of RT-AQF. Recent scientific advances in some of these areas that can lead to potentially more accurate RT-AQF are reviewed in Section 2. Major numerical, statistical, and computational techniques to further improve RT-AQF skills are described in Section 3. Section 4 presents several case studies to illustrate improved AQF skills and capabilities with advanced treatments and computational techniques. Major challenges and future prospects are provided in Section 5.

2. Scientific advances to improve air quality forecasts

2.1. Improvement of meteorological forecasts

Ambient pollutant concentrations depend on precursor emissions and meteorological conditions. Under typical emission scenarios, it is local-scale circulations and diurnal variations of meteorological variables such as solar intensity, temperature, wind speed, and mixing height that determine the daily and seasonal variations of chemical concentrations. Errors in meteorological forecasts are often readily propagated into RT-AQFs. Several studies reported a higher sensitivity of RT-AQFs to errors in the meteorological conditions, rather than in the emissions or chemical mechanisms (e.g., Hess et al., 2004). On the other hand, conventional meteorological approaches used in Numerical Weather Prediction (NWP) models were not designed for representations of air pollution episodes that typically have very weak dynamical forcing (Baklanov et al., 2002; Seaman, 2003); meteorological model improvements are, therefore, needed to support specific needs for RT-AQFs. Specific needs and challenges of meteorological modeling in support of air quality modeling and forecasting are summarized in several studies (e.g., Seaman and Michelson, 2000; Seaman, 2003; Dabberdt et al., 2004, 2006). For example, meteorological models do not handle some small-scale circulations (e.g., land-sea breeze, topographic induced circulations) at scales of $<$200 km and physical processes (e.g., turbulent mixing, PBL depth, cloudiness, precipitation and fluxes of heat, moisture, momentum, and mass) well. Most current meteorological measurements were designed for NWP, additional measurements (e.g., PBL height) under atmospheric conditions that are critical to air pollution episodes (not necessarily for NWP) would be very useful for validation and improvement of the parameterizations representing those conditions.

In addition, accurate meteorological model simulations at a horizontal grid resolution of $\sim$1 km or smaller are urgently needed to support RT-AQF in urban areas. This fine scaling modeling of meteorology poses challenges as many parameterizations were developed for larger scale applications. Improved parameterizations of physical processes are needed to more accurately represent stable/stagnation conditions, turbulence, deep convection, shallow clouds, low-level jets, nocturnal transport, land-surface processes, and representations of feedbacks among aerosols, clouds, and precipitation (Seaman, 2003; Dabberdt et al., 2006). The parameterization of urban areas within operational meso- and larger-scale models is particularly important but poorly represented. Significant efforts in improving parameterizations for urban meteorological modeling are ongoing and will be described in detail in Section 2.3.1. Detailed land-surface modeling that incorporates urban building structures and vegetative canopies have been formulated and tested (e.g., Masson, 2000; Martilli et al., 2003; Chen et al., 2004, 2011; Holt and Pullen, 2007; Miao et al., 2009). Such improved land-surface treatments will greatly improve the PBL representations and urban sub-layer flows (e.g., wind fields, thermodynamic structure, and turbulence) and consequently RT-AQF.

2.2. Improvement of chemical inputs

Inaccuracies in chemical inputs including ICONs and BCONs and emissions contribute to forecast errors. Chemical ICONs and BCONs can be derived using three methods: outputs from a global (or a synoptic scale) CTM, assumed climatological profiles, and adaptation of satellite and surface data for chemical profiles. Uncertainties and limitations exist for each method. The errors in a global CTM will cause errors in RT-AQF. The meteorological fields obtained from a global CTM are often at much coarser temporal (e.g., 6-h or longer) and spatial (e.g., $>1^\circ \times 1^\circ$) scales, therefore, an interpolation is needed to match the grid spacing for RT-AQF at urban/regional scales. Use of a global-through-urban model (e.g., GATOR-GCMOM, Jacobson, 2002; GEM-AQ, Neary et al., 2007; GU-WRF/Chem, Zhang et al., 2008, in review) with consistent model treatments can reduce forecast errors. Satellite measurements of vertical profiles of chemical species such as O_3, NO_2, NO_3, CO, SO_2, and HCHO, as well as aerosols could be used to improve the accuracy of the ICONs and BCONs. However, these satellite data are subject to uncertainties in the retrieval algorithms and inaccuracies due to cloud contamination and reflectivity of the surface. An accurate characterization of emissions of chemical species will directly improve model forecasting skills. Most RT-AQF models use offline emissions that are often generated based on historical emission patterns and do not account for variations under the current weather conditions. The use of online emissions that reflect real-time emissions and meteorological conditions will improve the accuracy of RT-AQF. These emissions may include those from online mobile sources, online biogenic sources, electric power generation; surface coating; wildfires corrected based on satellite data; dust events; sea-salt, and re-emissions from surfaces (e.g., NH_3, Hg). Additional techniques to improve accuracy of chemical ICONs, BCONs, and emissions using data assimilation and addressing their uncertainties using ensemble forecasting are described in Section 3.

2.3. Improvement of physical, dynamic, and chemical treatments

CTMs of RT-AQF systems include a large number of parameterizations for physical, dynamical, and chemical processes that govern the fates of air pollutants. The improvement of process treatments and parameterizations will yield long-term benefits to the improvement of the accuracy and efficiency of the RT-AQFs at various scales. Several excellent reviews of the modeling of those processes exist (e.g., Russell and Dennis, 2000; Zhang, 2008; Baklanov et al., 2011; Kukkonen et al., 2011) and we focus solely here on three main aspects that are particularly pertinent for RT-AQF: (1) the parameterization of the urban environment, (2) the gas-phase chemistry representation, which is essential for O_3, NO_2, and secondary PM predictions, and (3) aerosol dynamics and chemistry, which is relevant not only to PM predictions but also to O_3 and NO_2 predictions via the correct prediction of radiative transfer, photolysis and heterogeneous reactions.

2.3.1. Parameterizations for urban sublayer processes and physiographic data

Between 2011 and 2050, the world population is expected to increase by 2.4 billion, passing from 6.9 billion to 9.3 billion, and by 2030, 60% of the world population will live in cities (UNDESA, 2011). Given a rapid urbanization, the close linkage of RT-AQF to air pollution and associated human exposure in highly-populated cities, much of these model improvement efforts have focused on the parameterization of urban processes and the physiographic data that are critical for accurate RT-AQF. A wide range of micro- and mesoscale urban features can influence the atmospheric flow, its turbulence regime, the micro-climate, and, accordingly, modify

the transport, dispersion, and deposition of atmospheric pollutants within urban areas and in their vicinity downwind.

Incorporation of the urban effects into urban- and regional scale air quality models (AQMs) is generally carried out through improvements of meteorological fields (wind speed, temperature, turbulence, radiation, humidity, cloud water, precipitation) over urban areas. In comparison with NWP models, the urbanization for urban-scale RT-AQF models has specific requirements, e.g., requirements for a high resolution of the urban PBL vertical structure, because the correct surface fluxes over the urban canopy (UC) are not sufficient for urban-scale RT-AQF simulations. Accurate vertical profiles of the main meteorological fields and turbulence characteristics within the UC are needed to simulate the fate and impact of traffic emissions. Other important characteristics for pollutant turbulent mixing in urban-scale RT-AQF modeling is the mixing height, which has a strong specificity and heterogeneity over urban areas due to the internal boundary layer (BL) and blending heights from different urban roughness neighborhoods. The persistently increasing resolution in NWP models allows one to reproduce more realistically urban air flows and air pollution, and triggers interest in further experimental and theoretical studies in urban meteorology. Recent work performed by a consortium of FUMAPEX on integrated systems for forecasting urban meteorology and air pollution (Baklanov, 2006), and by the U.S. EPA and NCAR employing MM5 (Dupont et al., 2004; Taha, 2008a, b) and WRF (Chen et al., 2004, 2011), as well as other relevant work (Baklanov et al., 2008b, 2009), have disclosed many options and recommendations for the urbanization of NWP and meso-meteorological models. New local/street-scale RT-AQF systems with CFD models coupled with city- and mesoscale 3-D RT-AQF models are actively developed for industrial areas and megacities (e.g., San José et al., 2006, 2009; Baklanov and Nuterman, 2009). Such a forecasting downscaling system up to the street scale is being developed and tested for Copenhagen within the European MACC project (Baklanov and Nuterman, 2010).

Given different modeling objectives, there are three types of urbanization UC schemes: (1) single-layer and slab/bulk-type UC schemes, (2) multi-layer UC schemes, and (3) obstacle-resolved microscale models. The first two categories are sufficiently simple to be incorporated into operational atmospheric models. The third corresponds to computational fluid dynamic-type explicit building scale resolved models. The simplest approach is to modify the existing non-urban approaches (e.g., the Monin-Obukhov similarity theory (MOST)) for urban areas in an NWP model. Beginning with Brown and Wiliams (1998), who included urban effects in their turbulence closure scheme, methods with increasing levels of sophistication have been introduced into current mesoscale models. Masson (2000) included a detailed canyon energy balance scheme into the surface energy balance, whereas Dupont et al. (2004) included the effects from canyon walls, roofs, and streets in each prognostic PBL equation. A similar, but less complex urbanization scheme that shows promise toward capturing fine-scale urban weather phenomena, was a single-layer scheme developed by Kusaka and Kimura (2004a, b). These advances require detailed urban morphological data (i.e., on the scale of a few meters), including land use and land cover, surface roughness, building geometric and thermal characteristics, and anthropogenic heat fluxes (Ching et al., 2009). Thus, the next level of sophistication in NWP models may be achieved through implementation of advanced single- and multi-layer UC schemes, as it was implemented in Enviro-HIRLAM (Baklanov et al., 2008a) and WRF/Urban (Chen et al., 2011). This approach is a relatively inexpensive and practical means of improving on the modified MOST approach. For example, WRF/urban consists of a few options including a simple bulk parameterization, a single-layer UCM, and a sophisticated multi-layer UCM. It offers coupling to fine-scale CFD and large-eddy models for transport and dispersion applications, a capability of using high resolution urban land use, building morphology, and anthropogenic heating data using the National Urban Database and Access Portal Tool (NUDAPT) developed by Ching et al. (2009), and an urbanized high-resolution land data assimilation system.

Other specific features also affect air pollution in urban areas, but they cannot be realized via urbanization of NWP models and should be considered readily within CTMs. These features include (1) different pollutant deposition rates on specific urban surfaces, e.g., on vertical walls, due to different building materials and structure and vegetation; (2) chemical transformation specificities: increasing the chemical species lifetime due to subgrid-scale partitioning of air masses (e.g., inside street canyons), the heterogeneity of solar radiation (e.g., street canyon shadows) for photochemical reactions, specific aerosol dynamics, e.g., due to resuspension processes; (3) heterogeneity in sub-grid scale emissions of pollutants, especially due to traffic emissions, which needs to be simulated on a detailed urban road structure, considering traffic flow distribution; (4) pollutant indoor–outdoor interactions, which require more comprehensive emission modeling inventory; and (5) population exposure and air pollution adverse health effects, which are the final and most important aim of RT-AQF. It is, therefore, important to integrate RT-AQF and population exposure modeling, which includes high-resolution databases of urban morphology, population distribution and activities.

2.3.2. Other physical, dynamic, and chemical treatments

Although most current RT-AQF models have shown overall good/satisfactory skills in forecasting O_3 and $PM_{2.5}$ in terms of domain-average monthly/seasonal mean statistics, the comparisons of simulated and observed hourly O_3 and $PM_{2.5}$ values at individual sites show poor performance in capturing diurnal variations (e.g., underpredictions of daytime peak O_3 and overpredictions of nighttime O_3), spatial variation (large overpredictions or underpredictions of $PM_{2.5}$ at rural/national park sites), weekend vs. weekday trends (e.g., underpredictions of maximum 8-h average O_3 on weekends), and magnitudes at locations with special terrain/emission/meteorological characteristics (e.g., underpredictions in daytime O_3 at coastal urban locations) (e.g., Chen et al., 2008; Cai et al., 2008; Eder et al., 2009). While inaccuracies in meteorological forecasts and model inputs (in particular, emissions) undoubtedly contribute to such poor performance, several deficiencies in the model treatments of physical, dynamic, and chemical processes may contribute to such poor performance. Incorporation of recent advances into RT-AQF models will allow better representations of these processes, thus potentially improving the accuracy of RT-AQFs.

2.3.2.1. Chemical kinetic mechanisms. Several deficiencies and limitations in chemical kinetic mechanisms used in current RT-AQF models were recently identified. Luecken et al. (2008) found that SAPRC-99 predicted higher O_3 concentrations than CB05 and CB-IV. Faraji et al. (2008) attributed much of the difference between the SAPRC-99 and CB-IV mechanisms to differences in the chemistry of aromatics, especially mono-substituted aromatics such as toluene under NO_x-limited conditions. These studies indicated a large uncertainty in the representation of organic chemistry, in particular, aromatic chemistry, in current gas-phase mechanisms. Underpredictions of maximum O_3 concentrations are mainly due to the fact that CB05 and CB-IV mechanisms give lower O_3 production efficiency than observations. Kim et al. (2009) compared CB05 and RACM2 within Polyphemus and found that uncertainties in the kinetics of some major inorganic reactions (oxidation of NO by O_3 and HO_2) led to uncertainties commensurate with those due to the

organic chemistry (taking into account compensation of errors in the organic chemistry of the two mechanisms). Mollner et al. (2010) found that the uncertainty in the kinetics of HNO_3 formation (from the reaction of NO_2 with OH) could lead to differences of up to several ppb in O_3 concentrations. Kim et al. (2011a) in their comparison of CB05 and RACM2 over Europe showed that uncertainties in gas-phase chemistry impact SOA predictions because of uncertainties in oxidant concentrations and in the formulation of the organic chemistry (e.g., ring-conserving vs. ring-breaking oxidation pathways of the aromatic chemistry).

Using measured OH and HO_2 radical concentrations at an urban supersite in New York City, Cai et al. (2008) found that their RT-AQF model underpredicted O_3 production efficiency due to underpredicted odd hydrogen radicals (HO_x, the sum of $OH + HO_2$) in both summer and winter. Their study identified two additional deficiencies in most current chemical mechanisms used in RT-AQF models. First, the current gas-phase mechanisms, developed and tuned largely for summer conditions, cannot represent chemistry under winter conditions with low solar radiation and low temperatures. Mis-representation of HO_x chemistry in winter can lead to inaccurate forecasts of secondary PM formation. Second, the lack of a heterogeneous reaction of NO_2 on the surface of aerosols to produce HONO in the model is a main reason for the underpredicted OH levels. HONO photolyzes at sunrise to produce OH, which affects daytime O_3 production and atmospheric oxidizing capacity. A modeling study by Lei et al. (2004) showed that HONO formed through a heterogeneous reaction of NO_x on the surface of soot particles with a low limit of uptake coefficient can lead to 4–12 ppb increase with an average of 7 ppb in the daytime O_3 levels in the Houston-Galveston area. The occurrence of such heterogeneous reactions is supported with increasing evidence from field and laboratory studies (e.g., Su et al., 2008; Khalizov et al., 2010; Monge et al., 2010). A recent review of heterogeneous uptake and reactions on the surface of aerosols and clouds by Kolb et al. (2010) indicated the potential importance of a large number of heterogeneous chemical processes involving a number of trace gases (e.g., H_2O, OH, HO_2, O_3, NO_2, NO_3, N_2O_5, HONO, HNO_3, SO_2, chlorine nitrate ($ClONO_2$), bromine nitrate ($BrONO_2$), hydrochloric acid (HCl), and HCHO) on various types of surfaces including water surfaces, droplet surfaces, sea-salt, ice surfaces, mineral dust, soot surfaces, and solid and liquid organic surfaces as well as photochemistry on atmospheric surfaces such as soot and organic particles. At present, heterogeneous reactions other than heterogeneous hydrolysis of N_2O_5 are not included in most RT-AQF models, although some models such as Polyphemus include also heterogeneous reactions of HO_2, NO_3, and NO_2 on particles. These heterogeneous reactions can affect not only photochemical cycles but also sulfate and nitrate formation (Dentener et al., 1996; Zhang and Carmichael, 1999; Wang et al., in review). Another deficiency is a lack of gas-phase chemistry involving halogen species, which may be important in coastal urban areas. Recent field, laboratory, and modeling studies have shown that the chlorine radical (Cl) may enhance O_3 levels by several tens of ppb in coastal urban areas such as Houston (Tanaka et al., 2003; Chang and Allen, 2006; Sarwar and Bhave, 2007).

Several improved gas-phase chemical mechanisms accounting for some of the aforementioned deficiencies exist. For example, CB05 with Cl extensions for the troposphere (CB05Cl) includes 21 gas-phase chlorine reactions. It has been implemented into CMAQ and can increase O_3 mixing ratios by up to 8 ppb in the Houston area and 4 ppb in the New York–New Jersey area (Sarwar and Bhave, 2007). CB05 tends to underpredict the maximum O_3 and O_3 production rates under low-NO_x conditions. CB05 with a new toluene mechanism (CB05-TU) has been developed to address this limitation and it has been shown to perform better in predicting maximum O_3, O_3 formation rate, NO_x removal rate, and cresol concentration (Whitten et al., 2010). An updated version of CB05, i.e., CB6 with 77 species and 218 reactions, has been recently developed and shown to reduce underprediction bias in maximum O_3 as compared with CB05 (Yarwood et al., 2010). Although it is not used in most CTMs, a version of SAPRC-99 with 47 Cl reactions exists (Carter et al., 1997). SAPRC-99 has been recently updated to SAPRC-07 with 640 reactions among 222 species and a condensed version with 286 reactions among 84 species (Carter, 2010a, b). The major updates include new or improved representations of VOCs, reformulated aromatic chemistry and peroxy reactions for a better representation of SOA precursors, the addition of Cl chemistry based on Carter et al. (1997) and with additional reactions, and the updated rate constants and photolysis data. These updated gas-phase mechanisms will potentially improve both O_3 and PM forecasts.

2.3.2.2. Aerosol chemistry and dynamics. Inaccurate representations of aerosol chemistry and dynamics contribute significantly to the model biases in reproducing observed aerosol concentrations. Current RT-AQF models are unable to accurately predict the mass concentrations of SOA and to a lesser extent other volatile components such as NO_3^- and NH_4^+, the number concentrations and size distributions of PM, as well as the mixing state and radiative and hygroscopic properties of PM. Such inabilities severely limit the accuracy of PM forecasts and the possible extension of RT-AQF models to forecast other important variables that affect human health and climate change (e.g., mass/number concentrations and size distributions of nano-particles and aerosol radiative effects). More accurate representations of aerosol chemistry and dynamics based on up-to-date knowledge and research findings are needed.

Current regional and global AQMs significantly underestimate ambient OM, owing to incomplete treatments of SOA formation as well as uncertainties in the emissions of primary organic aerosol (POA) (and their atmospheric transformations) and gaseous precursors of SOA (e.g., Zhang et al., 2004; Yu et al., 2007; McKeen et al., 2007). Even with recent addition of several anthropogenic precursors (e.g., benzene, polycyclic aromatic hydrocarbons, and long-chain alkanes) and biogenic species (e.g., isoprene and sesquiterpenes) (Zhang et al., 2007; Couvidat and Seigneur, 2011), new SOA formation processes (e.g., in-cloud aqueous-phase oxidation of glyoxal and methylglyoxal and particle-phase oligomerization) (Pun and Seigneur, 2007; Carlton et al., 2010), and the refinement of the SOA formation pathways accounting for the effect of the NO_x regime (Carlton et al., 2010; Kim et al., 2011b), significant model underpredictions remain.

A recent breakthrough to this knowledge gap is the important contributions of SOA from gas/partitioning of POA and gas-phase oxidation of all low-volatility vapors generated from the evaporation of POA. A new volatility basis-set (VBS) SOA modeling approach (Robinson et al., 2007) was developed to efficiently treat SOA production from semi-volatile organic compounds, intermediate VOCs, and aged POA. A revised VBS approach that is based on volatility and oxidation state (i.e., the 2D-VBS) has also been developed; it tacks the evolution of OA and OA precursor gases to oxygenated organic aerosol through becoming increasingly oxidized, less volatile, and more hygroscopic during photochemical aging process (Jimenéz et al., 2009). Compared to the traditional two-product approach that requires two SOA surrogates for each reaction producing SOA, this new VBS approach has significantly improved the current model's efficiency in simulating SOA (Robinson et al., 2007; Murphy and Pandis, 2009; Shrivastava et al., 2010). The new VBS SOA module has been implemented into WRF/Chem v3.3. Similarly, SVOC emissions and transformation using the

hydrophobic/hydrophilic organic (H_2O) molecular approach have been incorporated into Polyphemus to provide similar improvements and considerably reduce discrepancies between model predictions and ambient measurements (Couvidat et al., 2011). Additional updates, e.g., the partitioning coefficients of SOA species, the volatility of POA, oxygen content, water solubility, and hygroscopicity of SOA should be made as they become available from laboratory measurements.

While current RT-AQF models focus on PM mass, the expansion of forecast products to include visibility, AOD, aerosol direct and indirect radiative forcing requires an accurate representation of PM number concentrations and size distributions, an inability that current RT-AQF models inherited from CTMs (e.g., Zhang et al., 2010a). The inaccuracy is largely due to inaccurate or missing mechanisms for new particle formation and subsequent growth processes. Various parameterizations for new particle formation have been proposed based on theories of binary (Kulmala et al., 1998; Yu, 2008), ternary (Merikanto et al., 2007; Yu, 2006), and ion-induced or ion-mediated nucleation (Yu, 2010), as well as limited observational data from laboratory/field studies (Sihto et al., 2006; Kuang et al., 2008). These parameterizations are being evaluated using increasingly available observed aerosol number and size distributions on regional and global scales. Recent studies showed substantial differences in the new particle formation rates calculated with different parameterizations and the resulting particle number concentrations (by up to 18 and 3 orders of magnitude, respectively) (e.g., Zhang et al., 2010a, b). Such discrepancies can be propagated into climate change predictions, leading to uncertainties in simulated aerosol direct and indirect effects (Makkonen et al., 2009). These studies also indicated a need to simulate early growth processes of newly formed particles before they grow into CCN size.

3. Techniques to improve air quality forecasts

A number of techniques have been developed to improve RT-AQF model performance in a shorter time frame, with a focus on the improvement of accuracy of the model inputs and forecast products. These methods are reviewed below.

3.1. Simple statistical techniques

3.1.1. Simple bias correction methods

Bias-correction techniques would be useful if they can correct model bias and improve RT-AQF effectively, although they cannot provide insights into model deficiencies or performance. Several simple bias correction methods have been implemented in a number of studies. The simplest and also most commonly-used method is the mean subtraction method, in which the mean bias is subtracted from the forecasted values at each monitoring site. This method, however, does not always guarantee a positive value. The second approach is the multiplicative ratio-adjustment method, in which the forecasted value is multiplied by the mean ratio of the sum of the observed value to the sum of the forecasted value at each monitoring site. This alternative correction guarantees that the concentrations will remain positive. McKeen et al. (2005) used the two simple bias correction algorithms for forecasts at each monitor location. They found that both methods reduced RMSE and increased RT-AQF skill, but the ratio-adjustment method provided additional improvement over the mean subtraction method for models with the highest biases. The third method is to force the zero differences between observed and simulated seasonal means by using an empirical linear fit between forecasted and bias-corrected values.

3.1.2. Data fusion methods

Data fusion is generally defined as the use of techniques that combine data from multiple sources to produce a single output that is more accurate and efficient than if they were achieved by means of a single source. Data sources may include observations, reanalysis data, and model data sets obtained from statistical models or 1-3D models. When a 1-3D model is used as one of the sources of data, one refers then to data assimilation. However, the combination of model outputs with observational data using kriging or interpolation techniques to create a reanalysis is sometimes referred to simply as data fusion. 3D RT-AQF models provide spatial and temporal details but often exhibit bias; on the other hand, a simple interpolation of monitoring data fails to take into account spatial and temporal dependencies present in the data. The data fusion methods have been used to overcome the limitations of RT-AQF models and observations by taking advantage of many years of community efforts in developing various statistical methods for RT-AQF, rapidly-matured advanced deterministic models, and increasingly available real-time observational data (e.g., AirNow or AQI indices) and combining them in a coherent way.

Post-simulation data fusion methods include various statistical methods that can help improve the forecasts when systematic deficiencies occur. For example, the Model Output Statistics (MOS) is a classical procedure in meteorology used to correct model forecasts at individual stations (Glahn and Lowry, 1972). In MOS, a site-dependent regression model, trained over past data, is applied to forecast parameters. Training seasons and implementation of the statistical MOS relies on a training procedure: observations over a past period and simultaneous forecasts are required. Compared with the raw forecast from CHIMERE, the MOS forecast reduced the RMSE from 19.5 $\mu g\ m^{-3}$ to 18.0 $\mu g\ m^{-3}$ on average for rural stations and improved the hit rate by 10%–16% (Rouïl et al., 2009). Guillas et al. (2008) used the Model Diagnostic and Correction (MDC) approach to downscale and improve local RT-AQFs using CTM outputs at 70 × 70 km and observed wind speed and precipitation from the U.S. EPA monitoring stations in the Atlanta area and showed that the statistically-adjusted outputs reduced forecast errors by up to 25%. Several advanced data fusion methods developed for improvement of retrospective air quality modeling can be adapted for RT-AQF. For example, Cowles and Zimmerman (2003) developed a Bayesian modeling approach for spatio-temporal data from two networks that account for possible differences in measurement errors, biases, and variances.

3.2. Advanced approaches based on chemical data assimilation

3.2.1. Data assimilation and its applications in RT-AQF

As a subset of data fusion, data assimilation refers to the process by which models and measurements are combined to produce an optimal representation of the state of the atmosphere. Data assimilation methods include nudging methods, statistical methods, variational methods (e.g., 3-dimensional variational method (3D-Var), 4D-Var), and sequential methods (e.g., optimal interpolation (OI), EKF, and EnKF). The types of observational data include those in-situ, airborne, and satellites. Data assimilation has been extensively used in the meteorological community, but its emergence in air quality modeling only dates back to the mid 1990s. For example, trace gas satellite data such as O_3, NO_2, and N_2O columns and vertical profiles of O_3, CH_4, HNO_3, $ClONO_2$, N_2O_5, N_2O, and CFC-11 were assimilated into stratospheric CTMs (e.g., Errera and Fonteyn, 2001) and tropospheric CTMs (e.g., Elbern et al., 1997) using various data assimilation approaches including OI and variational methods. OI was applied for the assimilation of AOD (e.g., Generoso et al., 2007) and NO_2 column mass (e.g., Wang et al., 2011). Wu et al. (2008) compared four assimilation methods for

assimilation of O_3 ground measurements: OI, EnKF, reduced-rank square root KF and 4D-Var. Satellite data assimilation has been particularly useful to improve stratospheric or upper-tropospheric O_3 (e.g., Eskes et al., 1999), boundary layer O_3 (e.g., Boisgontier et al., 2008), and sand and dust storm (Niu et al., 2008). Since the first applications of CDA for RT-AQF of Elbern and Schmidt (2001), CDA has been increasingly applied for RT-AQFs (e.g., Chai et al., 2006; Carmichael et al., 2008; Wu et al., 2008; Pagowski et al., 2010). Table 1 summarizes major approaches for CDA. A brief review of some of these approaches such as 4D-Var and EnKF can be found in Carmichael et al. (2008).

3.2.2. Overview of the methodologies based on CDA

The use of data assimilation techniques and its impact on the system's forecasting ability depend on the specific dynamics of the model. Global meteorological and oceanographic models have fewer forcings than AQMs: radiative, friction, and above all the ICON. For CTMs, the ICON is one forcing of many, less and less influential with time, whereas emission fields and sinks are strongly driving forcing fields. For limited area models and long-lived pollutants, BCONs may be as influential as emissions. Radiative forcings and species subsidence from the free troposphere are other forcings to account for.

Contrary to meteorology, the uncertainty attached to these forcing fields can be high. Emission uncertainty for usual air pollutants ranges from 20% to 60% (Hanna et al., 2001; J.-P. Fontelle, Centre Interprofessionnel Technique d'Études de la Pollution Atmosphérique (CITEPA), personal communications, 2010). Each of the data assimilation methods is different and has its own merits. For example, sequential methods rely on the assimilation of observations as they arrive to produce the best estimate of the chemical state of the atmosphere (Evensen, 2007). Differently, variational methods gather observations over a time period, and find the optimal model trajectory that accounts for these data, essentially following a least squares approach (Talagrand, 1997). Both methods have been developed and tested in the late 90's and early 00's on actual regional air quality applications (Segers et al., 2000; Elbern and Schmidt, 2001).

Although the air quality data assimilation community has inherited from meteorological advances, it has its own history and specific developments. For air quality studies, one can distinguish several types of application. Typical problems are the regulated air quality species: O_3 and precursors, VOCs, sulfate, PM, and OA with RT-AQF implementations; or the industrial/natural accidental atmospheric tracers (radionuclides, volcanic ashes). The mathematical techniques implemented to improve the ICONs are described in this section. The application of data assimimilation to the retrieval of parameter fields (i.e., inverse modeling) will be described in Section 3.3. But as mentioned earlier, because they represent major driving forces, this retrieval could also be an integrated component of the forecasting algorithm.

3.2.3. Sequential methods

Sequential methods are based on the Best Linear Unbiased Estimator (BLUE), that allows to find the best compromise between a set of observations and prior (or background) information on the system state, such as a previous forecast. This estimation represents the analysis step of the algorithm. It assumes a linear or linearized observation operator. For BLUE, it assumes a linear observation operator. Yet the assumption can be alleviated by using a linearized version of the operator, or a variational scheme (e.g., 3D-Var). It also relies on second-order moments closure of all statistics. A stronger but common assumption is that the statistics of the errors (observation, background) are Gaussian. In between analyses, the information on both the optimal state and possibly the uncertainty statistics on this state are propagated.

Examples of sequential methods include OI, KF, and many variants/extensions of the KF such as the EnKF. OI is a method that propagates the best estimate, but does not propagate any statistical information (Daley, 1993). It is coupled to a BLUE analysis. Statistical information must be provided from other source than the model forecast, such as climatological statistics on errors. OI is sometimes called kriging of innovation (Blond and Vautard, 2004). Although it is less and less considered in meteorology, it is still of importance for air quality applications because it does not require high computational resources and is yet efficient (Wu et al., 2008). Because of its lesser sophistication, it is also quite easy to implement and is, therefore, used to test data assimilation on new species (Tombette et al., 2009).

The KF is a self-consistent extension of BLUE that propagates the best estimate as well as the error covariance matrix that describes the uncertainty about it. It is defined for linear observation and model operators. Its non-linear extension, the extended Kalman filter (EKF) is based on the linearized versions of these operators. As a major drawback of the method, the state error covariance matrix cannot be stored for large applications, because it contains $N(N+1)/2$ degrees of freedom if N is the dimension of the state vector. Beside, the propagation of the matrix would require $2N$ model runs. That is why the direct use of the KF, or EKF is restricted to small to moderate size applications. The reduced-rank square

Table 1
Major techniques for chemical data assimilation.

Category	Subcategory	Strength	Limitation
Sequential method	Optimal interpolation	Very simple and robust, only optimal state estimation	Does not propagate errors
	Kalman Filter	Simple, propagate errors	Handles linear systems only, too costly for large systems
	Extended Kalman Filter (EKF)	Handles non-linearity	Uses the model tangent linear which is not robust, too costly for large systems
	Ensemble Kalman Filter (EnKF)	Handles non-linearity, no model adjoint, trivial parallelization of ensemble propagation	Sampling/reduction errors, needs inflation, localization
	Particle filter	Full Bayesian and non-Gaussian treatment of errors, no model adjoint, trivial parallelization of ensemble propagation	Collapse of the ensemble
Variational method	3D-Var	As simple as OI, handle non- Gaussian observation operator	Does not propagate errors
	4D-Var	Smoothing within the assimilation window, handles non-linearities, full (two-ways) propagation of errors, handles parameter estimation	Requires model adjoint, does not allow easy access to posterior errors, parallelization not as simple as EnKF; difficulty in the computation of the adjoint due to the high nonlinearities; may give negative concentrations and emissions

root filter method is useful since the use of KF cannot be contemplated without the reduction of the matrix of error covariance that needs to be propagated. Since error covariance matrices are bilinear quantities it can be shown that the KF can be formulated considering the propagation of the columns of the square root of such a positive definite matrix. Besides, the analysis step can also be "factorized" in terms of these modes. Then the reduction step consists in keeping only a limited group of $r << N$ biggest modes, of high uncertainty, that will be propagated later on in the forecast step. These modes are often augmented with additional modes that represent other source of errors, such as model error, so prominent in air quality. The reduced-rank square root filter has been applied to air quality in several studies (e.g., Segers et al., 2000; Hanea et al., 2004; Wu et al., 2008).

The EnKF (Evensen, 1994) pushes the sampling concept of the reduced square root filter one step further, making it a Monte Carlo scheme. An ensemble of state vectors is used to generate error statistics, through the empirical mean and error covariance matrix of the ensemble. These empirical moments are used in the BLUE analysis at the analysis step. In the stochastic version of the filter, there is an analysis for each member of the ensemble, with the same gain matrix for all, but a differently perturbed observation vector for each member (Burgers et al., 1998). This is meant to account for the proper statistics. During the forecast step, all members are propagated by the model (possibly non-linear). This is one major advantage because no linearization is necessary and no bias is thus introduced. The forecast statistics are recomposed later using the propagated ensemble. The EnKF and its variants are now a method of choice in geophysical data assimilation, but it has weaknesses that are shared with the other filters.

Several issues are common for the sequential methods, namely, inflation, localization, and model error. First, the sampling of the errors statistics with the ensemble is only an approximation of the true statistics. It leads to an underestimation of the errors that needs to be compensated by an ad hoc inflation (Anderson, 2001) of the errors statistics. The ensemble dispersion is artificially increased to avoid any overconfidence that could lead to filter divergence. These errors can be estimated online and adaptively (Constantinescu et al., 2006a). Errors can also be modeled through a perturbation scheme that is meant to represent un-identified errors. Because of many uncertain forcings, this generation of errors has been emphasized in air quality data assimilation (Constantinescu et al., 2006a; Wu et al., 2008). Model errors, including emission, BCONs, kinetic rates error, are so important in air quality that the inflation operation needs to be implemented via stochastic perturbations of each member of the ensemble. Each perturbation is a linear combination of identified sources or errors. Their variability is often represented by log-normal distributions given the nature of the uncertain variable (positive quantity). The finer these sources of model errors are diagnosed, the better the filter is expected to perform. In addition, the sampling schemes can generate long-range spurious correlations that need to be removed. In AQMs, it can generate unphysical blobs of pollutant in the domain. That is why localization schemes were created. One solution is to apply a short distance correlation function to the estimation of the error covariance matrix via a Schur product that guarantees that the resulting matrix is still a covariance matrix (Hamill et al., 2001). Another way is for each grid cell of the domain to assimilate observations only locally (Houtekamer and Mitchell, 1998). Both methods yield very satisfying localization. However it introduces ad hoc parameters that tune this localization, such as a correlation length. Finding the optimal parameters is a currently active field of research (for inflation and localization). Localization has been used in CDA only recently (Eben et al., 2005; Constantinescu et al., 2006b).

There are many other sequential methods (e.g., Singular Evolutive Extended KF (SEEK), Singular Evolutive Interpolated KF (SEIK), unscented filter, Ensemble Transform KF (ETKF), Local Ensemble Transform KF (LETKF)). Most of them are similar to the filters mentioned above, with similar performance. Choosing one of them is less of an issue than in meteorology where model errors are less intrusive, making the precise choice of the algorithm a real issue. A few of them have been specifically applied to photochemistry (Hanea et al., 2004). Particle filters are sequential data assimilation techniques that apply the Bayes formula, from which the BLUE analysis can be derived under the Gaussian hypothesis (which is thus not needed any more). Because of a filter divergence (at the analysis step, the particles tend to collapse onto a single one), it is difficult to implement them on high-dimensional geophysical problems. The interest of particle filters for RT-AQF is not demonstrated, and a matter of debate, since even though the physics may be very non-linear leading to non-Gaussian distribution of errors, the dynamics is essentially not chaotic (Bocquet et al., 2010).

3.2.4. Variational methods

BLUE can be replaced with a 3D-Var analysis, which, for a linear observation operator is equivalent. Therefore, the analysis step can be made variational (leading to the minimization of a cost function, also referred to as objective function, in place of the application of a linear algebra formula) with a natural and exact extension to a non-linear observation operator. But such a variational approach can also be carried out for the forecast step. The optimization operates on a time window (typically 24 to 48-h). A least squares cost function is designed to compute the mismatch between the model trajectory and the observation, and the ICON and prior information on this condition (which could be climatological or resulting from a previous forecast). Because time is now accounted for in the window, the variational method deals with 4-D fields, hence its name 4D-Var (Le Dimet and Talagrand, 1986). This is easy to write but enforcing the fact that the 4-D field of concentration is a model trajectory is difficult and related to optimal control theory. The minimization of this cost function requires the computation of the adjoint of the tangent linear of the forecast model (in addition to the observation operator). This can be a formidable task for complex geophysical numerical models, even though automatic differentiation software can partially help in that matter. This is the main drawback of the variational approach, which is often compensated by its benefits (no loss of information within the window, optimal treatment of non-linear operators). However, 4D-Var has been successfully implemented operationally in meteorology, giving a decisive advantage to the centers operating the method, even though it has been shown recently that the EnKF could compete (Buehner et al., 2010). 4D-Var has been tested and implemented quite early in air quality (Elbern and Schmidt, 1999, 2001), and is also used in the operational EURAD RT-AQF model in Cologne.

When the retrieval of emission parameters is not taken into account, Wu et al. (2008) have shown that OI and EnKF perform better than 4D-Var. This is because 4D-Var does not account for model error. This could be achieved by using the so-called weak-constraint 4D-Var that allows for model errors. This is, however, very close to a 4D-Var that would allow retrieving not only an initial state but also model parameters. Indeed, 4D-Var lends itself to the optimization of parameters along with the initial state condition. It was recommended early to optimize a selection of parameters of the emission field for an optimal forecast (Elbern et al., 2000). This is why much more than in other geophysical fields, data assimilation in air quality cannot be separated from the inverse modeling of parameters. Difficulties come from the specification of background

inter-species error covariance for concentrations at the initial time or emissions over the whole time window. It also comes from the computation of the adjoint because of the high nonlinearities in the chemistry, in the aerosol evolution and/or the thermodynamics related to multiphasic microphysics.

3.3. Inverse modeling using data assimilation

Different from ICONs, the impact of uncertainties in other model inputs such as emissions and BCONs and model parameters such as chemical reaction kinetic rate constants will persist throughout the simulation. Beyond the estimation of the chemical state of the atmosphere, the retrieval of the forcing fields, such as emissions, BCONs or model parameters, has its own interest. For emissions, this is the so-called top-down approach: improving inventories through observations and a CTM. Previous studies have demonstrated the effectiveness of using various inverse modeling methods to obtain the spatial distribution of pollution emissions as well as to improve the forecast skill of AQMs (e.g., Xu et al., 2008). For large fields, 4D-Var is the method of choice. Its feasibility on real emission factor of NO_x was proven by Quélo et al. (2006). The large-scale inversions of the precursors of O_3, SO_2, SO_4^{2-}, NH_3, VOCs, etc., were successfully carried out by Elbern et al. (2007) over Europe. The inventories were corrected and were supporting the general belief on the economical growth of eastern Europe and the emission control over western Europe. Hakami et al. (2005) estimated BC emissions over East Asia using the adjoint STEM model. Yumimoto and Uno (2006) applied 4D-Var to a CTM and estimated CO emissions over East Asia. Koohkan and Bocquet (submitted for publication) also applied 4D-Var to estimate CO emissions, but over France and simultaneously using a statistical sub-grid model to counteract strong representativeness errors. As a long-term forecasting tool, the system lead to a strong reduction of the bias, a high correlation of more that 70%, when traditional CO regional CTM simulations do not usually exceed 30%. Recent developments and results of chemical adjoint are presented by Henze et al. (2007). In another example, a factor in the dust emission parameterization scheme was estimated using the lidar vertical profiles of dust particles (Yumimoto et al., 2007). Underestimates in the dust emission fluxes were corrected and more reasonable dust concentrations were achieved.

For linear models, or model amenable to linear effective models, the direct use of the BLUE analysis can be carried out to constrain the BCONs with observations. Roustan and Bocquet (2006) carried this approach for gaseous Hg over Europe, implicitly correcting the northern BCON, thereby reflecting the influence of Hg depletion events. When a few parameters are to be estimated, 4D-Var might be inappropriate, and stochastic filters are usually efficient. For instance, Barbu et al. (2009) estimated the kinetic rate of conversion of SO_2 to SO_4^{2-} in Europe. Using a multi-purpose 4D-Var, Bocquet (in press) has simultaneously optimized the emission, the Kz and Kh parameters, and several micro-physical paramaters (e.g., dry deposition velocity, wet scavenging ratio). Applied to a tracer dispersion event (Chernobyl), it leads to a strong reduction of biases, and of the representativeness and model errors (80% correlation in validation).

The retrieval of parameters is however a difficult task because it requires specifying the uncertainty of parameters that are often very different in nature. The uncertainties are themselves poorly known. Besides, their response function may be very non-linear, and the optimization is numerically difficult with an outcome of uneasy interpretation. Another difficulty of inverse modeling and data assimilation in atmospheric chemistry is the positivity of concentrations and emissions (e.g., Constantinescu et al., 2006a). Indeed since data assimilation techniques are based on the BLUE paradigm, they assume Gaussian errors, which contradicts the positivity of the related variables (Bocquet et al., 2010). An unappealing but widespread solution, which may often suffice, is to set negative concentrations or emission fluxes to zero. Another possibility is to use non-Gaussian error priors based on positive distributions. Either via a Bayesian or Maximum Entropy on the Mean inference, they lead to non-quadratic cost-functions for variational data assimilation (Bocquet, 2005a, b, 2008). By constraining the system even more, this usually results in better retrievals. The methodology has been successfully applied to the ETEX I and II, Algeciras, and Chernobyl dispersion events (Bocquet, 2007; Krysta and Bocquet, 2007; Davoine and Bocquet, 2007). It has been applied to the estimation of the cesium-137 and iodine-131 Fukushima Daiichi accident source terms (Winiarek et al., 2012), with a mathematically rigorous estimation of the uncertainty of the retrievals.

Multiscale data assimilation methods can be used to complement some limitations in observations. At high resolution, the number of variables of the emission field is very large compared to the number of observations. Besides, because of the dispersive nature of atmospheric transport, the information content of the observations is not shedding light on all grid cells, especially those away from the monitoring network. An idea is to define a relevant adaptive grid for the emission fields that take into account the network and the meteorology for an optimal assimilation of observations (Bocquet, 2009; Bocquet et al., 2011). The adaptive emission field grid is coarse in the regions with no influence on the observations, whereas the grid is refined close to the stations to the highest available resolution. Another method is the optimizing the monitoring network for an optimal forecast. This method and the targeting of sondes in optimal locations for observations have been proven useful in meteorology. However the global observational system is very dense and the gain is relatively limited. In air quality, with a still limited impact of satellite data, a sparser ground network, and higher representativeness errors, optimal design of the networks for nowcasting of O_3 fields (Nychka and Saltzman, 1998; Wu et al., 2010; Wu and Bocquet, 2011), or targeting of observations for forecasting a contaminant plume (Abida and Bocquet, 2009), have been proven quite useful.

3.4. Advanced approaches based on ensemble forecasting

3.4.1. Ensemble forecasting and its applications in AQMs

Ensemble forecasting is a numerical prediction method that is used to produce a representative sample of the possible future states of a model system. Ensemble forecasting can be implemented using multiple models or one model but with different inputs (e.g., varying meteorological input forcings, emission scenarios, chemical ICONs) or different process parameters (e.g., varying chemical reaction rates) or different model configurations (e.g., varying grid spacings). Since their first application to one CTM with three different meteorological inputs by Vautard et al. (2001), ensemble techniques have been applied to many RT-AQF systems. The work of Delle Monache et al. (2004) represents one of the first efforts for the multi-model real-time O_3 forecasts. Similar to the improvement found for O_3 ensemble forecasting (e.g., McKeen et al., 2005; Pagowski et al., 2005; Delle Monache et al., 2006a), ensembles of the $PM_{2.5}$ forecasts showed significant statistical improvements over any individual forecast (McKeen et al., 2007). A possible reason for such improvements is that ensemble averaging removes part of the unpredictable components of the physical and chemical processes involved in O_3 and PM formation, as compared with any deterministic ensemble member. However, very few applications addressed uncertainty quantification or probabilistic forecasts based on ensembles of forecasts (e.g., Delle Monache et al., 2006b, 2008; Vautard et al., 2009; Garaud and Mallet,

2011). The resulting ensemble forecast is usually a weighted linear combination of the individual ensemble members. Inherent limitations are associated with individual ensemble members. Accuracy may be sensitive to the weighting factors used. Most methods compute the ensemble forecast at the observed locations only. Some combination methods (e.g., sequential aggregation) may not account for observational errors. More sophisticated approaches aimed at improving ensemble forecasting have been developed. One such method is the ensemble forecast of analyses (EFA) of Mallet (2010). EFA couples an ensemble forecasting approach (i.e., sequential aggregation) with CDA techniques to forecast an analysis from data assimilation, instead of observations; it performs well with 28% reduction in RMSE of surface O_3 forecasts as compared to a reference simulation.

3.4.2. Overview of the methodologies based on ensemble forecasting

AQMs are limited by large uncertainties in their physical and numerical formulations, and their input data. As a result of the multi-scale nature of the modeled phenomena, the models rely on approximate sub-grid parameterizations that introduce errors on the mean concentration fields. The computational costs may be so high that only simplified chemical mechanisms and aerosol dynamics can be considered. Such simplifications are also attributed to the lack of appropriate data. The numerical discretization is another source for errors, e.g., because of rather coarse model grid cells, or because of the limited number of aerosol size sections. A CTM relies on many uncertain input fields whose errors propagate to all components of the CTM's state. Following Hanna et al. (1998, 2001), one can assume that the meteorological fields are available with $\pm$20% uncertainty or more, that the anthropogenic emissions can show higher uncertainty than $\pm$50%, and that the biogenic emissions are known only within a factor of two. Many other data (e.g., BCONs, land use cover) are subject to significant errors. In this context, it is doubtful that picking a single model with a single set of input data is the best strategy for forecasting. Instead, a stochastic standpoint let the concentrations be considered as random variables with some joint probability distribution. Ensemble approaches were developed so that all the uncertainty sources can be taken into account in the forecasting process. Several forecasts are produced by various numerical models that rely on different physical formulations and input data sets. The generation of an ensemble based on a single model with perturbed input data is referred to as Monte Carlo simulations. When the ensemble relies on different physical and numerical models, with or without perturbations in the input data, the approach is referred to as a multi-model approach. If the ensemble properly represents the uncertainties, it can sample the probability distribution of the output concentrations or at least estimate the uncertainty of its forecasts. The empirical variance of the ensemble provides a confidence interval along with the forecast itself (which may be given by an ensemble mean). If the uncertainty description is accurate enough (which is seldom the case), an application is forecasting the exceedance of some regulatory threshold. This activity can turn into probabilistic forecasting when the objective is forecasting the probability that a given event occurs.

In all ensemble applications, a limiting factor is the computational cost. The generation of an ensemble of size N may require N times the computational resources of one model simulation. The actual cost can be lower if the simulation system is able to share certain demanding operations (in pre-processing) between different simulations. However, the cost remains high, especially in a forecasting context. In practice, 10-100 simulations can reasonably be carried out on modern forecasting platforms. The size of the ensemble may therefore not be large enough in order to properly approximate the probability density function of the models' state (which usually contains one to ten million components). Table 2 summarizes major ensemble forecasting techniques.

3.4.3. Monte Carlo (MC) simulations

MC simulations consist in carrying out a number of simulations with perturbed input data. The perturbations are sampled according to probability distributions chosen by the modeler, and the perturbations associated with two simulations should be independent (in the basic MC approach). As the number of simulations increases, it is possible to obtain an approximation of the mean, standard deviation, and even probability distribution of the output concentrations. The mean of the simulations' concentrations converges to the expectation of the concentrations, with a rate independent of the dimension (i.e., number of chemical species and model's grid cells) but at a slow rate (i.e., with a convergence rate proportional to the square root of the number of simulations). One clear advantage toward MC simulations is their simplicity, since they solely require the ability to run simulations based on independently perturbed input data. Hanna et al. (1998) carried out Monte Carlo simulations for O_3 over New York City during three days in 1988. They perturbed 109 parameters of the model UAM-IV based on the probability density functions (normal or log-normal) that were advised by ten experts. A similar study was carried out by Hanna et al. (2001) over the eastern U.S. in order to evaluate the impact of emission control strategies. Beekmann and Derognat (2003) and Deguillaume et al. (2008) applied the MC approach over the Paris area and computed a posteriori uncertainty estimates with a Bayesian approach. Recent work tried to better describe the

Table 2
Major techniques for ensemble forecasting.

Category	Strength	Limitation
Monte Carlo simulations	Monte Carlo simulations are reasonably easy to implement and their mathematical framework is well known.	The uncertainties due to the model physical and numerical formulations are essentially ignored.
Multimodel ensembles	The ensemble takes into account the uncertainty in the models' formulation. It can be combined with Monte Carlo simulations. The variety of the models can bring a lot of information.	The approach may be difficult to implement in practice. The number of models may be low when the models are provided by different teams.
Sequential aggregation	Strong improvements can be expected from the most advanced methods. These methods are robust, fast and easy to implement. They require tuning of few parameters.	They do not always take into account observational errors, and the spatial distribution of the weights may not be reliable. Usually, the forecasts for non-observed species cannot be improved.
Coupled sequential aggregation and classical data assimilation	It overcomes the limitations of sequential aggregation without loss of performance.	The method requires an ensemble and the application of a data assimilation method, which are not available on every operational platform.

uncertainties in the input fields with spatialized perturbations (Boynard et al., 2011). Spatialized perturbations can soften or prevent the covariance localization issue, which is especially important in a data assimilation context. Perturbing the meteorological fields raises difficulties related to the consistency among the perturbed fields. A better option is the use of meteorological ensemble forecasts as input to the CTMs (e.g., Straume et al., 1998; Straume, 2001).

3.4.4. Multimodel ensembles

Multimodel ensembles are more complex to build since they involve CTMs based on different physical and numerical formulations. Contrary to MC simulations where the shape of the perturbations is an explicit parameter, the differences between two models from a multi-model ensemble are difficult to anticipate and control. Nevertheless, these differences address what Pinder et al. (2009) call structural uncertainty that cannot be represented with MC simulations alone. The members of the ensemble can be either models developed by different teams (e.g., McKeen et al., 2005; van Loon et al., 2007) or models built within the same numerical platform (e.g., Mallet et al., 2007). The former case leads to ensembles with typically 4-10 models. Such an approach was carried out for uncertainty estimation for photochemistry (Delle Monache et al., 2006a; Vautard et al., 2009). In the case of models built on the same platform, the platform should allow flexible changes in the physical and numerical formulations (Mallet and Sportisse, 2006). This strategy makes it possible to generate large ensembles and to fully control the design of the ensemble (Garaud and Mallet, 2010). Garaud and Mallet (2012) showed that the MC approach fails to generate a member with higher spatio-temporal variability than the observations because the model lacks variability and this is not compensated by the perturbations in the input data. On the contrary, the multi-model ensemble contains several models whose variability is greater than that of the observations. It is possible to combine a multi-model and MC approach, so as to take into account all uncertainty sources. Pinder et al. (2009) proposed an intermediate strategy where the uncertainty due to inputs is approximated with the sensitivity to these data, which allows to generate large ensembles at lower cost.

3.4.5. Ensembles calibrated for uncertainty estimation

An important concern with ensemble simulations is the accuracy of their representation of the uncertainties. Besides an a priori knowledge of the model performance, the main additional source of information lies in the observations. Hence scores involving observations were introduced to measure the quality of an ensemble, in terms of uncertainty estimation. We assume the ensemble contains N members. The most common score is based on the rank histogram. It checks whether the N members of the ensemble properly sample the concentrations (seen as random variables). It is primarily used to check that an uncertainty estimate (in practice, an empirical standard deviation) is reliable. For each observation, the corresponding ensemble concentrations are sorted in increasing order, and a rank among the sorted concentrations is given to the observation. The rank is 0 when the observation is lower than all concentrations, and is N when the observation is greater than any concentration. The rank histogram displays the number of observations per rank. If the ensemble is well balanced, the rank histogram should be flat. If not, the shape of the histogram reveals the deficiency of the ensemble. For instance, it is common to obtain a U-shaped histogram which reveals an underestimation of the uncertainty—with many observations outside the envelop of the ensemble.

In order to measure the quality of a probabilistic forecast, one usually evaluates: (1) the reliability which relates to the consistency between observed occurrence frequencies and probabilistic forecasts, (2) the resolution which is the ability to produce significantly different probabilistic forecasts for some subsets of events (which the climatological forecast cannot achieve), and (3) the sharpness which measures the ability to forecast extreme events. The reliability diagram and the Brier score are two classical tools. They are always computed for a given event, and they assess how efficiently the ensemble produces the probability of the event to occur. The reliability diagram plots the observed occurrence frequency of an event against the forecast probability of this event. If the ensemble is reliable and if a large number of occurrences are observed, the observed frequency should be the same as the forecast probability. Hence a perfect reliability diagram should coincide with the first diagonal. An ensemble with no skill will lead to a flat diagram since, for any forecast probability, the mean occurrence (i.e., climatological) frequency of the event is observed. The Brier score is the mean quadratic error of probabilistic forecasts. For each date i, the ensemble forecasts a probability p_i, between 0 and 1, and the observed "probability" o_i is either 0 if the event did not occur, or 1 otherwise. The quadratic error at that date i is given by $(p_i - o_i)^2$. Because of the uncertainties, the probability p_i cannot be exactly 0 or 1 at all dates. Hence the Brier score cannot be zero, even if the ensemble accurately estimates the model uncertainty. This makes the score difficult to interpret, but the Brier score provides an important reference when comparing the abilities of two ensembles.

The ensemble verification scores serve as objectives in the design of an efficient ensemble. In Monte Carlo simulations, one may adjust the uncertainty levels associated with the input data; with multi-model ensembles, one may include more models with further changes in their formulation. However, the optimization of the ensemble can hardly be automatic: besides the algorithmic difficulty, the cost of ensemble simulations is so high that an optimization loop (involving the generation of a new ensemble at each iteration) can barely be an option. There is however a need for an automatic calibration of ensembles for reliable uncertainty estimation. Daily RT-AQF is especially demanding since an ensemble well balanced over a large period of time may fail on a shorter period. One option is to adjust the uncertainty estimates with statistical methods. Another option developed by Garaud and Mallet (2011) consists in generating a large ensemble that preferably overestimates the uncertainties, and to select a sub-ensemble that performs well according to one of the aforementioned scores. This approach allows one to extract a 20 to 30-member sub-ensemble out of a 100-member ensemble, and it allows relying on an ensemble re-calibrated before every forecast.

3.4.6. Sequential aggregation

In order to produce a single improved forecast out of an ensemble, a few methods were proposed in recent years. The simplest methods are the ensemble median and the ensemble mean. The former was essentially applied for passive tracers (Riccio et al., 2007). The second was applied for photochemical applications (Delle Monache and Stull, 2003; McKeen et al., 2005; van Loon et al., 2007). In these cases, the ensemble mean showed some improvement in the forecasts (though not for all scores), but there is no guarantee that the ensemble mean will improve the forecast in any case—and in several unpublished cases, the ensemble mean actually showed lower performance than the best model in the ensemble.

In order to better use the information provided by the ensemble, weighted linear combinations of models were developed. Before every forecast, the weights are computed based on past observations and past and present model simulations. The forecasts are then linearly combined with the chosen weights, which produces

a single aggregated forecast, hopefully more efficient than the forecast of any member of the ensemble. This process is repeated sequentially before each forecast. We therefore refer to this approach as sequential aggregation. Following the super-ensemble approach of Krishnamurti et al. (2000), least-square methods were applied by Mallet and Sportisse (2006). The weights of the linear combination are determined so as to minimize the mean quadratic discrepancy with observations over a learning period. The weights have a limited validity in time, which requires that they be constantly recomputed over a (moving) learning period of typically 30 days before the forecast. Despite the practical efficiency of the approach, there is little theoretical support and guarantees as of the performance.

Pagowski et al. (2006) applied regression to determine the weights of the linear combination. In order to adapt the weights in time and not to require large data sets, they applied the dynamic linear regression (West and Harrison, 1999), which allows the characteristics of the random processes to be time-dependent. This approach proved to be efficient for O_3 forecast over southern Canada and the eastern U.S. during 56 summer days. A robust approach relying on machine learning methods was proposed by Mallet et al. (2009). The weights can be computed with methods such as the exponentiated gradient (for convex combinations) or the ridge regression (for non-convex combinations) (Cesa-Bianchi and Lugosi, 2006) in discounted versions. The main advantage of these methods is their mathematical framework that guarantees, in the long run, a performance (compared to the observations) at least as good as that of the best constant (in time) linear combination. This guarantee holds whatever the sequence of observations and models' forecasts may be, which makes the method very well adapted to operational forecasts. They have been applied for several years on the operational platform Prev'air (see Section 4.2). A drawback of this approach is that the observation errors are not taken into account.

3.4.7. Coupled sequential aggregation and classical data assimilation

The main drawback of the previous aggregation methods is their lack of support for multivariate fields. The weights are computed per monitoring station and observed species. In the best cases, the weights are learned at all observed locations at once, so that they should have some spatial validity. A solution is to forecast the analyses (generated by some data assimilation methods) instead of the observations (Mallet, 2010). The analyses are supposed to be the best a posteriori knowledge of the true atmosphere state, and they are multivariate fields. A machine learning algorithm can be employed to forecast these analyses, with weights computed independently for every grid cell and chemical species. This approach guarantees that, in the long run, the aggregated fields forecast the analyses at least as well as the linear combination of models' forecasts with the best constant (in time, but space- and species-dependent) weights. Even if the weights are computed independently in every grid cell, the approach is able to capture all mean patterns found in the analyses. Since summer 2010, this ensemble method is operationally used for the Prev'air platform.

4. Case studies

4.1. RT-AQF with WRF/Chem-MADRID and WRF-CMAQ

WRF/Chem-MADRID of Zhang et al. (2010b) was deployed for RT-AQF at a horizontal grid resolution of 12-km in the southeastern U.S. Its forecasting skill in terms of spatial distribution, temporal variation, and domainwide statistics is evaluated using available U.S. EPA AirNow hourly observations for August 3–9, 2008 and intercompared with that of the U.S. NOAA's WRF-CMAQ of Eder et al. (2009). There are a number of differences between the two model systems. First, WRF/Chem-MADRID is an online-coupled meteorology and chemistry model, whereas WRF/CMAQ version 4.6 is an offline-coupled model. Second, WRF/Chem-MADRID uses the Advanced Research WRF (ARW) dynamic core of WRF (WRF/ARW), which is different from the nonhydrostatic mesoscale model (NMM) dynamic core (WRF/NMM) used in WRF/CMAQ. The physical configurations for WRF/NMM are based on those used in Eta for simulating cloud microphysics, boundary layer, surface layer, cumulus parameterizations, and longwave and shortwave radiation without cloud effect on the optical depth. They are different from those used in WRF/Chem-MADRID. Third, although both models use the same CB05, they use different aqueous-phase chemistry, the CMU bulk aqueous-phase mechanism in WRF/Chem-MADRID and a modified RADM aqueous-phase chemistry in WRF/CMAQv4.6. Fourth, WRF/Chem-MADRID uses the MADRID 1 aerosol module of Zhang et al. (2010a, b) which represents the particle size distribution with eight size sections over the PM aerodynamic diameter range of 0.0215–10.0 μm. WRF/CMAQ uses the aerosol module AERO4 that represents the particle size distribution with three lognormally-distributed modes for the size range of 0.001–10 μm. They both use ISORROPIA v1.7 to simulate the thermodynamic equilibrium of inorganic PM, but differ in the treatments of SOA formation and PM dynamic processes such as nucleation, condensation, and coagulation. For example, WRF/CMAQ utilizes a SOA module that accounts for SOA formation from 8 classes of condensable SVOCs, 6 from anthropogenic precursor VOCs (from 3 classes of aromatics including xylene, toluene, and cresol, and 1 class of higher alkanes) and 2 from biogenic monoterpenes. In WRF/Chem-MADRID, the SOA module includes 7 classes of condensable SVOCs from anthropogenic VOCs (including high-yield aromatic species such as toluene, ethylbenzene, ethyltoluenes, and n-propylbenzene; and low-yield aromatic species such as xylenes, trimethylbenzenes, dimethylethylbenzenes and tetramethylbenzenes) and 25 classes of BVOCs (from 18 BVOCs including monoterpenes, sequiterpenes and isoprene).

Fig. 1 compares 6-day mean simulated concentrations of maximum 8-h average O_3 and 24-h average $PM_{2.5}$ and OM from both models and available AirNow observations. The performance statistics for meteorological and chemical predictions are summarized in Tables 3 and 4. Both models give similar daily maximum 8-h average O_3 mixing ratios in terms of spatial distribution, magnitude, and performance statistics, with a slightly better statistical performance for WRF/CMAQ. The moderate overprediction of O_3 by both models is mainly due to inaccurate biogenic and point source emissions. Other possible explanations include overpredictions of temperature at 2-m (T2) and underprediction of wind speed at 10-m WS10 (see Table 3), as well as possible inaccuracies in simulated PBL height. Different meteorological predictions by WRF/ARW and WRF/NMM are partially responsible for differences between WRF/Chem-MADRID and WRF/CMAQ simulations, in addition to their differences in chemistry and aerosol treatments. WRF/Chem-MADRID gives slightly higher biases for WS10 but lower biases for T2 and total precipitation. It predicts a daytime PBL height that is lower by 12% domainwide than that of WRF/CMAQ, leading to slightly higher O_3 mixing ratios and slightly worse A, CSI, POD, and FAR, as compared with WRF/CMAQ.

For 24-h average $PM_{2.5}$, WRF/Chem-MADRID performs better than WRF/CMAQ in terms of both statistics (Table 4) and spatial distributions (Fig. 1). For example, daily absolute MBs on August 7–9 are <1 μg m^{-3} for WRF/Chem-MADRID but ~ 3 μg m^{-3} for WRF/CMAQ. Both models underpredict $PM_{2.5}$ concentrations during the 6-day time period, due likely to overpredictions in precipitation and T2, incorrect biogenic emissions, uncertainties in

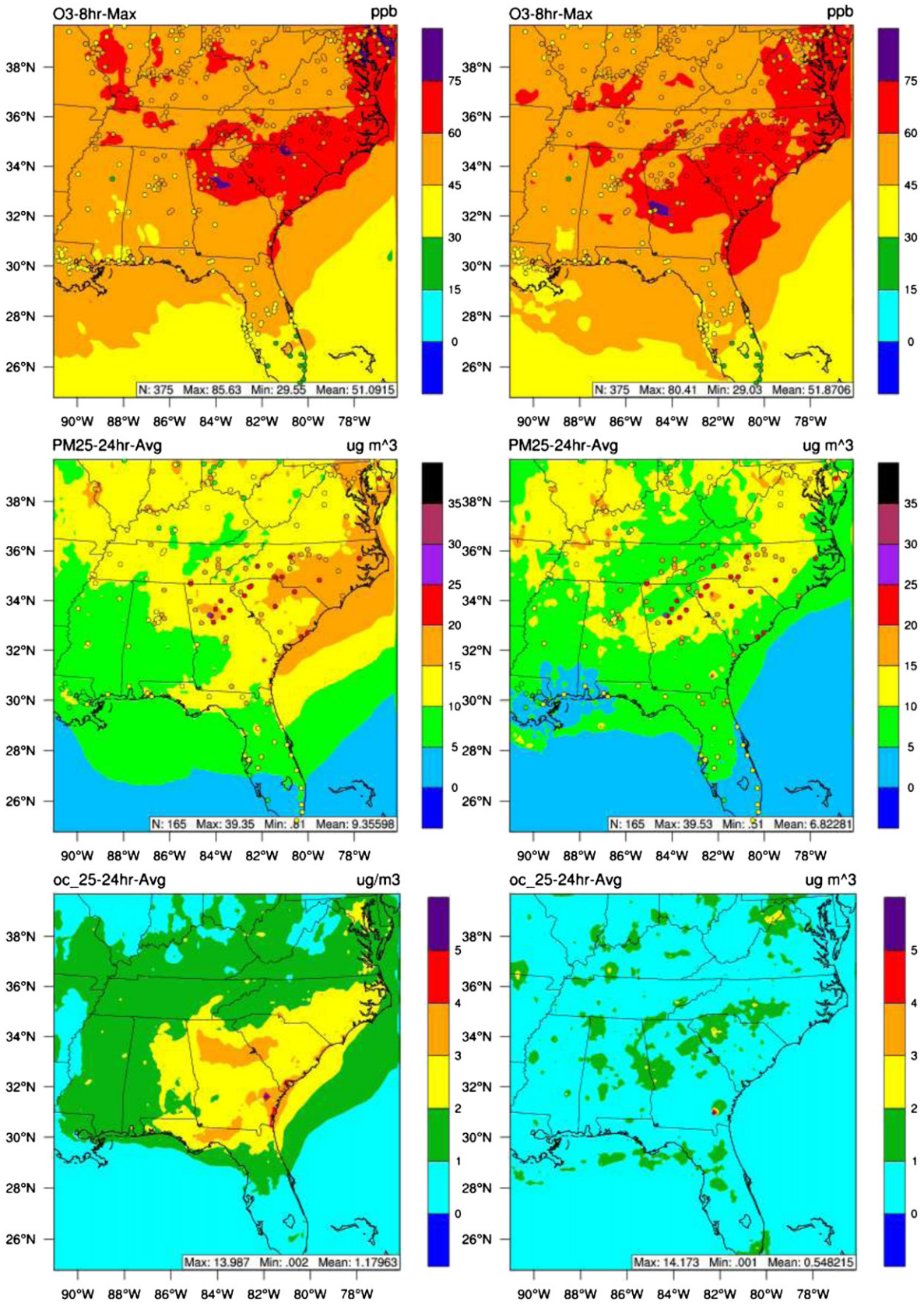

Fig. 1. Overlay plot of six-day (August 4–9, 2008) mean concentrations of maximum 8-h average O_3 (top), 24-h average $PM_{2.5}$ (middle), and 24-h average OM (bottom) for WRF/Chem-MADRID (left) and WRF/CMAQ (right) (circles indicate observations from AIRNow, http://www.epa.gov/airnow, no observations of OM were available from AIRNow).

Table 3
Performance statistics evaluation of meteorological forecasts for August 4–9, 2008.

	Hourly T2 (°C)		Hourly WS10 ($m\ s^{-1}$)		Daily total precipitation ($mm\ day^{-1}$)	
	WRF/Chem- MADRID	WRF/CMAQ	WRF/Chem- MADRID	WRF/CMAQ	WRF/Chem- MADRID	WRF/CMAQ
Mean Obs	25.2	25.2	4.5	4.5	1.4	1.4
Mean Sim	25.3	25.8	3.1	3.6	1.8	2.1
MB	0.1	0.6	−1.4	−0.9	0.4	0.7
RMSE	2.1	2.5	3.0	3.1	10.1	6.6
NMB (%)	0.5	2.2	−31.2	−19.6	26.3	47.2
NME (%)	6.0	7.3	43.2	43.4	209.5	187.2

T2: Temperature at 2-m; WS10: Wind Speed at 10-m. Obs: Observation; Sim: Simulation; MB: Mean Bias; RMSE: Root Mean Square Error; NMB: Normalized Mean Bias; NME: Normalized Mean Error.

emissions of primary PM and PM precursors such as NH_3 and VOCs, as well as incomplete model treatments such as SOA formation from glyoxal. Both models give similar spatial distributions of SO_4^{2-}, NH_4^+, other inorganics, and EC, but WRF/Chem-MADRID tends to give higher values due likely to lower PBL height and less over-prediction of precipitation. Much larger differences exist for OC (see Fig. 1), NO_3^-, and Na^+ between the two models. Several studies have reported underpredictions in OC due to missing precursors/processes in CMAQ (e.g., Tesche et al., 2006; Zhang et al., 2007). Missing of some SOA species due to incorrect biogenic emissions and SOA formation from sesquiterpenes and isoprene can explain in part the low OC predictions from WRF/CMAQ. By contrast, MADRID treats additional SOA formation from sesquiterpenes and isoprene, leading to higher OC values and better agreement with observed $PM_{2.5}$. WRF/CMAQ distributes sea-salt emissions into the coarse mode, whereas WRF/Chem-MADRID simulates sea-salt emissions online that produces sea-salt in both fine and coarse modes, leading to higher $PM_{2.5}$ concentrations over oceanic areas than those simulated by WRF/CMAQ. The inclusion of reaction between sea-salt and HNO_3 in WRF/Chem-MADRID, which is not considered in WRF/CMAQ, may affect NO_3^- predictions in coastal areas. When WS10 is strong along the coast and over the ocean, emitted sodium chloride (NaCl) can react with HNO_3 to form sodium nitrate ($NaNO_3$). The equilibrium approach is used to simulate gas/particle partitioning in WRF/Chem-MADRID, which leads to high NO_3^- in the fine PM size range (Zhang et al., 2010b). These results show an overall improved $PM_{2.5}$ forecast with more detailed model treatments.

4.2. Ensemble modeling with sequential aggregation and coupling with data assimilation

Ensemble forecasting has proven to be helpful in RT-AQF by Prev'air. This platform is operated by the French Institut National de l'EnviRonnement industriel et des rISques (INERIS). Every day, it forecasts hourly concentrations for O_3, NO_2, $PM_{2.5}$, and PM_{10}, over France, for the current day and the next two days. Several AQM simulations, among which five with aerosols, are carried out over Europe, and six simulations are carried out at the global scale ($2^\circ \times 2^\circ$ resolution), over Europe ($0.5^\circ \times 0.5^\circ$ resolution), or France ($0.15^\circ \times 0.1^\circ$ or $0.1^\circ \times 0.1^\circ$ resolution). The simulations use CHIMERE (Schmidt et al., 2001), MOCAGE (Peuch et al., 1999), and a configuration of Polair3D from Polyphemus (Mallet et al., 2007). These simulations define over France (but with different resolutions) an eight-member ensemble for gases and a five-member ensemble for PM. Analyzing the ensemble led to the conclusion that it can be efficiently exploited for ensemble forecasting, despite its small size. After a test period in 2008 (Debry et al., 2009), sequential aggregation has been applied since 2009 to these ensembles in order to produce daily ensemble forecasts on the platform.

In 2009, a first version of ensemble forecasting relied solely on machine learning algorithms (i.e., sequential aggregation), following Mallet et al. (2009). The performances of the models were mainly measured with correlation, bias and RMSE, using hourly observations from about 150 French stations representative of the simulation spatial resolution (i.e., near-source stations were not included). After the night simulations, the weights were computed based on all hourly observations and corresponding simulated concentrations from the previous days. The discounted ridge regression was employed to provide weights for the current day and the next two days, which is a fast operation since learning algorithms have low computational requirements. Compared to the model with the lowest RMSE, the RMSE of the ensemble forecast was typically 15%–30% lower. For peak O_3, the lowest model RMSE was 19.8 $\mu g\ m^{-3}$ while the RMSE of the ensemble forecast was 16.7 $\mu g\ m^{-3}$ Fig. 2 shows the daily O_3 concentration profile, averaged over all monitoring stations and for summer 2009, for the aggregated forecast as well as for the eight individual forecasts on the operational platform Prev'air. A near-perfect agreement was obtained using ensemble forecast.

As of 2010, the ensemble approach coupled with data assimilation is employed to produce the ensemble forecast. The analysis is

Table 4
Performance statistics of chemical forecasts for August 4–9, 2008.

	Maximum 1-h O_3 (ppb)		Maximum 8-h average O_3 (ppb)		24-h average $PM_{2.5}$ ($\mu g\ m^{-3}$)	
	WRF/Chem- MADRID	WRF/CMAQ	WRF/Chem- MADRID	WRF/CMAQ	WRF/Chem- MADRID	WRF/CMAQ
Mean Obs	54.8	54.8	48.7	48.7	14.8	14.8
Mean Sim	62.6	61.8	56.8	56.4	12.6	10.6
MB	7.8	7.0	8.1	7.7	−2.2	−4.2
RMSE	14.6	13.6	13.2	12.2	6.7	7.3
NMB (%)	14.3	12.7	16.7	15.8	−15.1	−29.0
NME (%)	20.3	18.4	20.9	19.3	35.9	39.7
A (%)	89.7	90.9	72.3	76.3	65.1	64.2
CSI (%)	17.1	23.7	35.7	39.4	36.0	24.7
POD (%)	47.1	64.0	85.2	88.0	46.6	28.4
B	2.2	2.3	2.2	2.1	0.8	0.4
FAR (%)	78.7	72.6	61.9	58.3	38.8	34.6

Obs: Observation; Sim: Simulation; MB: Mean Bias; RMSE: Root Mean Square Error; NMB: Normalized Mean Bias; NME: Normalized Mean Error; A: Accuracy; CSI: Critical Success index; POD: Probability Of Detection; B: Bias; FAR: False Alarm Ratio.

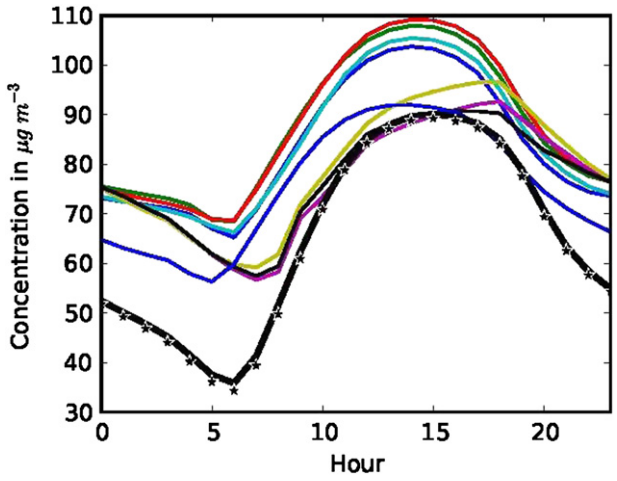

Fig. 2. Daily O_3 concentration profile, averaged over all monitoring stations and during summer 2009, for the aggregated forecast as well as for the eight individual forecasts on the operational platform Prev'air. The values of the aggregated forecasts are plotted with a thick black line; the observations are marked with the stars; and the other lines are for eight individual models available during this period (taken from Debry and Mallet, in preparation).

computed for hourly and daily O_3 with the kriging of the discrepancy between a reference simulation and the observations. According to É. Debry (INERIS, France, personal communications, 2010), RMSE for the current day is reduced by 18% for hourly O_3 concentrations (from 19 μg m^{-3} for the best model to 15.6 μg m^{-3}), and by 27% for daily O_3 values (from 17.8 μg m^{-3} for the best model to 12.9 μg m^{-3}). Note that the best model for hourly values has an RMSE as high as 23.4 μg m^{-3} on daily O_3; hence the ensemble forecast reduces the error by 45% compared to this model. On the Prev'air platform and in non-operational studies, the EFA ensemble forecasting brought larger improvements than the other methods (ensemble-based or assimilation-based), in terms of statistics and retrieval of spatial patterns. However, it does not or slightly increases the performance of the forecasts of threshold exceedances.

4.3. FUMAPEX urban air quality information and forecasting system (UAQIFS)

Urban and street air pollution is usually higher than pollution in rural areas and associated with significant adverse health effects. Prediction of health effects and implementation of urban air quality information and abatement systems require accurate simulation of air pollution episodes and population exposure on city and street levels, as well as the indoor–outdoor relationship of the pollutants. As a result, RT-AQF needs to consider a multi-scale modeling approach for urban areas with downscaling by different models from meso- and city-scale with parameterizations of sub-grid urban effects to the local- and micro-scales. However, most of the current models fail to produce realistic meteorological fields for the urban-scale RT-AQF. The EU project FUMAPEX developed a novel approach that integrates the latest developments in meteorological, air quality, and population exposure modeling into UAQIFS. Fig. 3 shows the application of an improved UAQIFS for the London metropolitan area (Sokhi et al., 2006). The improved city-scale MM5 integrated with the urban air pollution modeling system, in comparison with the original non-urban version, showed a substantial improvement of the RT-AQF for urban areas: differences between predicted and observed values can be reduced by up to a factor of three by using the high resolution urban land cover characteristics.

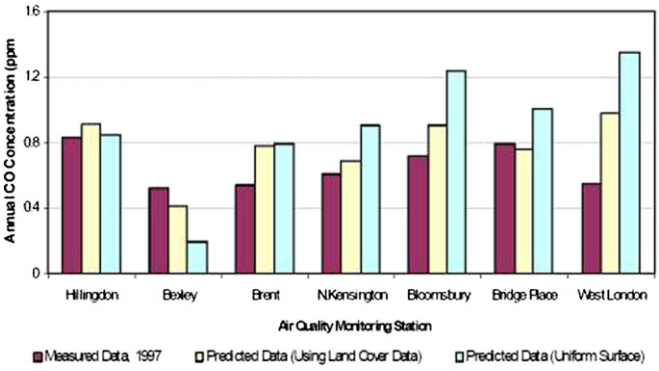

Fig. 3. Comparison of predicted annual CO concentrations (ppm), using the original non-urban version (right columns) and improved city-scale MM5 model integrated with the urban air pollution modelling system (central columns), in comparison with measured values (left columns) from seven monitoring stations in London (Sokhi et al., 2006).

5. Major challenges and future prospects

5.1. Summary of advances in sciences and computational technology

Addressing model deficiencies/limitations, improving scientific treatments, and incorporating recent and emerging advances based on up-to-date knowledge are key to the improvement of accuracy of RT-AQF. Recent scientific advances in urban parameterizations, gas-phase chemistry and aerosol chemistry and dynamics can potentially improve the accuracy of RT-AQF. A number of numerical, statistical, and computational techniques have been developed with a focus on improving inputs and forecast products. These include various bias adjustment techniques to correct biases in forecast products, various CDA techniques for improving chemical ICONs and emissions, and various ensemble forecasting approaches to quantify the uncertainties of the forecasts or to improve the forecasts. There is a growing trend to combine some of these approaches (e.g., the KF predictor bias-corrected ensemble forecasts, EFA) to achieve optimal accuracy. RT-AQF provides extraordinary opportunities for applications of advanced mathematical, statistical, and computational techniques. Continuous development efforts on those techniques will significantly reduce forecasting biases and yield immediate benefits in accuracy.

5.2. Current limitations, major challenges, and future directions

3D RT-AQF models have provided a powerful tool for RT-AQF but there remain some limitations in many aspects of the models. Addressing these aspects poses significant challenges for RT-AQF, as highlighted below.

5.2.1. Challenges in improving accuracy of meteorological forecasting

The most challenging meteorological forecasting in support of RT-AQF lies in weather patterns with weak synoptic, dynamical forcing (e.g., high pressure systems, calm winds, stagnant conditions). The major challenges include:

- Improve many existing parameterizations in meteorological models that were previously designed for large-scale applications or develop new parameterizations for finer scale applications. These may include schemes to represent PBL, radiation, turbulence, dispersion, deep and moist convection, shallow clouds, precipitation, land-surface processes, and sea-breeze circulation.

- Improve meteorological observing systems by including air quality-related variables (e.g., PBL height, vertical profiles of temperature, specific humidity, and wind speed) in routine networks, make more continuous real-time satellite/sounding data. Couple the improved models with improved observing systems through implementation of various data assimilation and modeling approaches to improve the model performance.
- Conduct sensitivity simulations of historic episodes using different combinations of physical options for major processes and nudging options to identify an optimal combination that may achieve the best model performance for RT-AQF over a specific domain and time period.
- Develop and improve online-coupled meteorology and chemistry models to accurately represent their interactions and feedbacks, in particular, the feedbacks among aerosol-cloud-precipitation. Key processes that need attentions include aerosol activation/resuspension, nucleation and auto-conversion of cloud droplets to rain droplets, in-cloud and below-cloud scavenging, the effect of aerosols on ice crystal formation, and aerosol and cloud micro-physical effects on convection and precipitation, and convection-microphysics feedbacks.

5.2.2. Challenges in improving accuracy of RT-AQF model inputs

More efficient approaches are needed to generate inputs as accurately and quickly as possible and to adjust them with technically-sound approaches efficiently due to the time pressure of RT-AQF. The major challenges include:

- Develop more reliable instrumentations for measurements of emissions (e.g., continuous emission monitoring) and the integration of the field/laboratory measurements into online emission modules.
- Develop and improve online emission modules needed to simulate weather-dependent emissions such as emissions of BVOCs, dust, sea salt, VOCs from surface coating, and re-emitted species (e.g., NH_3) from various surfaces (e.g., soils, vegetations, and water), and wildfire emissions.
- Develop and improve methods for inversion modeling with CDA (e.g., 4D-var) to better estimate primary emissions of pollutants (e.g., SO_2, NO_x, CO, NH_3). While some methods do exist, areas with difficulties remain, e.g., the retrieval of parameters affecting emissions, the qualification of the associated uncertainties, the nonlinearity of the chemical system, and the estimation of the real emissions instead of numerical adjustments compensating for errors in other inputs.
- Reduce uncertainties in ICONs and BCONs through more frequent adaptation of observations from surface networks, sounding, and spaceborne sensors by using CDA that continuously ingests real-time observations or efficient bias correction methods based on previous day's observations.
- Develop global-through-urban online-coupled models to provide dynamic ICONs and BCONs for nested simulations at small scales to reduce uncertainties caused by inconsistencies in the model treatments between offline-coupled global and urban/regional models.

5.2.3. Challenges in improving representations of urbanization

A reliable urban-scale forecast of meteorological and chemical fields is critical to urban emergency management systems for air-quality warnings, accidental toxic releases, fires, and chemical, radioactive, or biological substance releases due to terrorist actions. The potential risk of these real and frightening emergency episodes has recently emerged as homeland security issues. An accurate representation of urban structure, meteorology, chemistry, and heat island effect, remains a key to the improvement of RT-AQF at such fine scales. Major challenges include:

- Improve major urban parameterizations and schemes including those for atmospheric chemistry and specific aerosol dynamics due to heterogeneity of emissions and solar radiation inside street canyons, the urban PBL structure, the estimations of urban energy balance, urban vegetation, sub-grid cloud treatments, as well as the snow treatments.
- Improve treatments for subgrid-scale representations of local concentrations in urban areas by including accurate representations of local emissions, dispersion, and chemistry processes.
- Improve urban characterizations by increasing pointwise measurements within the urban canopy for parameterization validation and by using high-resolution, remotely-sensed data to derive urban parameters (e.g., land use, surface albedo, emissivity, and heterogeneity).
- Utilize CFD models to develop and improve urban parameterizations to enhance the understanding of the most important mechanisms of the urban atmospheric canopy layer and urban roughness sub-layer. Develop and improve online mobile source emission inventories (i.e., dynamic traffic models coupled with instantaneous or near-instantaneous emission factors) for incorporation into RT-AQMs. These models could provide the most accurate emissions at a scale of a few meters, thus greatly improving the capability of the forecasting skill in urban areas.
- Develop/improve human exposure models to simulate the neighborhood-scale population exposure to major air pollutants and forecast environmental health. Such exposure modeling establishes quantitative linkages between air pollution and epidemiological impacts at urban levels and represents a very important direction of practical usage of RT-AQFs.

5.2.4. Challenges in improving representations of other processes and aspects

Forecasting errors and biases are often caused by inaccuracies and uncertainties in model representations of chemical and physical processes of air pollutants. Other sources of errors such as uncertainties and inconsistencies in real-time observations obtained with various platforms may also contribute to forecasting errors and biases. Major challenges include:

- Establish/expand comprehensive real-time chemical measurement systems to better characterize abundance, spatial and temporal distributions, and physical and chemical properties of pollutants and source–receptor relationships. Improve data management and quality assurance systems to ensure proper collection and distribution and near real-time access of observations for RT-AQFs.
- Improve measurement techniques to characterize micro-physical properties of clouds and aerosols (e.g., cloud droplet number, PM number, mass, and size distributions and hygroscopicity, PM and cloud optical properties) in urban areas with very high concentrations of cloud droplets and aerosols. Improve representations of VOC chemistry via integrated field/laboratory measurements and 3-D modeling to accurately simulate O_3, SOA, and organic air toxics such as HCHO. Reduce the uncertainties in the kinetics of major inorganic chemical reactions via well-designed laboratory experiments using advanced measurement techniques.

- Develop and improve accurate modules for PM thermodynamics and dynamics, which are crucial to accurate RT-AQF. These include modules for new particle formation, SOA formation, and gas/particle mass transfer. Improve numerical techniques for aerosol micro-physical simulations in terms of both accuracy and computational efficiency. Processes consuming most CPUs include coagulation, dynamic gas/particle mass transfer, calculation of activity coefficients of organic species, organic and inorganic PM interactions, fine and coarse PM interactions, aerosol production via aqueous-phase chemistry, and aerosol activation by cloud droplets.
- Couple RT-AQF models with improved observational systems via CDA techniques and various bias correction methods to improve next day's forecasting.
- Conduct sensitivity simulations of historic episodes using different combinations of options/mechanisms to identify an optimal mechanism combination with the best model performance for RT-AQF over a specific domain and time period. This approach is subject to the same constraints (e.g., large metrics of mechanisms/schemes, limited computer resources, and knowledge of the users) as that for sensitivity simulations with an NWP model. An additional challenge is that the best physical option combinations identified through sensitivity simulations of meteorological forecasting models may not necessarily lead to the best performance due to intricate relationship between meteorology and chemistry.
- Develop and improve online-coupled meteorological and RT-AQF models to enable an accurate representation of the interactions and feedbacks among meteorology, reactive gases, and PM in the real atmosphere. Develop global-through-urban online-coupled models to provide consistent chemical and physical treatments for nested simulations at urban-to-regional scales. Such consistencies can help reduce model errors/uncertainties in forecasting at small scales.
- Apply a well-designed coupler to couple various model components (e.g., between atmospheric and land processes) and minimize time required for communications and data exchanges.

5.2.5. Challenges in applying techniques for accuracy improvements

Many numerical and computational approaches have demonstrated promising skills in reducing forecasting errors. Continuous development and improvement of these techniques are critical to the refinement of the forecasting skills of RT-AQF at all scales. Major challenges include:

- Develop various bias correction approaches to minimize model biases. These approaches largely rely on statistical methods and available observations. Current applications of these approaches focus on ground-level forecasting products; expanding such techniques to correct vertical profiles of chemical species, column variables, and top BCONs will be technically challenging. A skillful utilization of the state-of-the-art statistical methods and limited observational data is required to develop effective bias correction techniques.
- Improve/expand various CDA techniques to reduce uncertainties in model inputs and inaccuracies in existing and new forecasting products including concentrations, deposition fluxes, and their spatial and temporal variabilities, as well as species vertical profiles and column abundance. Variational methods (e.g., 3D-Var and 4D-Var) have certain advantages over other methods in handling nonlinearity of the chemical system and some observational data such as infrared radiance data from satellites and should be further developed. The major challenge remains a better understanding and control over errors that may be intrinsic or extrinsic to the scheme.
- Develop and apply ensemble forecasting methods to reduce forecast errors and estimate the resulting uncertainties. Designing meaningful members and size of ensemble simulations with limited computational resources and evaluating the relative benefits obtained with multi-model vs. single-model ensembles are difficult tasks. Probabilistic forecasting can better describe the nature of likely events than purely deterministic forecasting that often contains inherent and unquantified uncertainties. It represents the most challenging aspect of ensembles in which the ensembles must be designed with a proper representativeness of uncertainties using carefully-selected probability distribution of the model output.
- Develop and apply post-simulation data fusion methods (e.g., MOS) to reduce model errors through a combination of observations, a statistical model, and 3-D RT-AQF predictions. The main challenges are to obtain sufficient amount of observations for data training to develop a robust linear regression model and apply this model skillfully to correct predictions at individual sites made by the deterministic model. Another challenge lies in the implementation of advanced statistical methods such as the Bayesian modeling approach into this data fusion framework.

5.2.6. Challenges in applying techniques for computational efficiency improvements

Some approaches have been developed in the past to address the computational aspects of RT-AQF. These techniques have demonstrated promising skills in handling the computation-related issues including memory requirement, efficiency, stability, and scalability. Their continuous development and improvement will undoubtedly support the continuous success of RT-AQF at all scales and expansions of its applications over more areas by more users, especially, those who cannot afford high performance computing clusters. Major challenges include:

- Develop efficient parameterizations/representations of the detailed model treatments to be used in the operational RT-AQF and a benchmark to assess the magnitude of the potential biases introduced by simplified treatments and the impact on the forecasting products.
- Ensure a high level of parallelism for computer codes for an optimal speedup. Parallel computing has been increasingly used for RT-AQFs on various platforms and should be continuously used. Parallel computer programs, however, are more difficult to write than sequential ones, because concurrency, communication, and synchronization between different tasks and subtasks often create the greatest obstacles to achieving an optimal performance.
- Implement a high level of automation for RT-AQF in all steps involved in forecasting including data downloading, pre-processing, model simulations, data exchange between grids and model components, post-processing and analysis, and web-posting.
- Use refinement or enrichment techniques such as the dynamic adaptive grid techniques for multi-scale nesting to surmount the numerical difficulties associated with nested grids by a continuous grid. These techniques can resolve important features and allocate computational resources more wisely to the resolution of scales needed for a better forecast and also bridge the gaps between local and mesoscales and between meso- and global scales more efficiently.

- Explore other approaches for their potential in improving efficiency yet maintaining a satisfactory accuracy. These may include identifying the most time-consuming components for optimization, using different numerical approaches to solve atmospheric processes more efficiently, using more efficient numerical solvers, developing regime-dependent chemical mechanisms with fewer reactions and species for certain areas/subdomains to reduce computational demands, optimizing model configurations and forecasting simulation design on the available computer architectures, and tailoring the model performance toward the operational supercomputing hardware and the parallel computing environment for effectively managing input and output processes.

5.2.7. Challenges in promoting community outreach and increase of awareness

Since RT-AQF is now a common practice at all levels of government and research organizations worldwide and the RT-AQF products are being widely used to issue health advisories and alert the public to pollution episodes, RT-AQF has several unique challenges in technical training, community outreach, and increase of public awareness that are typically not encountered (or to a much lesser extent) by conventional air quality modeling. Major challenges include:

- Develop forecast guidance documents. Such guidance should cover the methods, tools, and models to be used for RT-AQFs, the evaluation protocols to judge their performance, and the methods to interpret and use forecasting products. They can also identify areas of improvements and priority research areas to guide proper allocations of limited resources and serve to nucleate immediate attention of the research and operational communities for pressing issues.
- Establish effective education and training programs and develop material to train potential forecasters at all levels to meet the RT-AQF needs. Increasing amounts of resources should be allocated from various governmental agencies, educational organizations, and private sectors to educators from research organizations and universities to develop and maintain a long-term university curriculum and training programs to train the next generation of air quality forecasters.
- Develop and strengthen community outreach and public awareness programs. Adequate resources should be allocated from governmental agencies to implement and support such programs to disseminate forecasts and health concerns and outreach college/university students, K-12 teachers and students, public and private stakeholders (e.g., researchers and educators), health-affected population, economy-affected sectors, societal and economic impacts specialists, health insurance specialists, farmers and other agricultural producers, educational and other medias (weather service providers, broadcast meteorologists, Internet providers, newspapers, radio, mobile services), and the emergency-response community. Such programs should include forecast dissemination, needs assessment, education, communication, and value assessment.
- Support and coordinate centralized RT-AQF efforts region-, country-, and worldwide and promote domestic and international science, information, data exchange, and experience and knowledge sharing. Air pollution occurs at all scales and some become regional and global concerns that are not limited to geographical boundaries of the regions and countries. Systematic coordination and close collaborations among multiple organizations/countries to consolidate limited resources for advancement of RT-AQF are important to its continuous success.

5.3. Concluding remarks and outlooks

Similar to weather forecasting that affects everyone, RT-AQF has emerged as a new forecasting discipline and is undergoing substantial and rapid advances. It intersects with many science and engineering disciplines but stands out among other disciplines with a special societal demand and technical requirement uniqueness. It represents one of the most far-reaching developments and practical applications of science and engineering, poses unprecedented technical and computational challenges, and provides significant opportunities for science dissemination, community communications, and societal participations to outreach a variety of stakeholders. In the past decades, many cities and countries worldwide have successfully launched RT-AQF systems that are based on tools and models with varying degrees of sophistications ranging from the simplest rule of thumb to the most advanced 3D online-coupled meteorology and chemistry models.

For the decades to come, we envision a new generation of a comprehensive RT-AQF system that will be centered on the state of the science 3D RT-AQF models and supplemented with sophistical statistical models. This system will be equipped with many modern technologies to reduce forecasting biases and enhance computational efficiencies including advanced techniques for multi-scale data assimilation, multi-model ensemble forecasting, adaptive downscaling, data fusion, and statistical post-processing and be supported with a suite of real-time or near real-time observational data from all platforms. The 3-D RT-AQF models are now in the process of transition from offline-coupled to online-coupled models and from mesoscale models to unified systems across scales from global to urban. Such an advanced and comprehensive RT-AQF system will address multiple air pollution issues and resulting impacts at multiple scales (e.g., local-to-urban air pollution, long-range transport, adverse health effects) and will be capable of forecasting multiple pollutants (e.g., O_3, PM, CO, VOCs, NH_3, acids, and air toxics) on a short term basis (on the order of a few to 10 days). This system can also be extended to forecast air quality and its impacts in a long-term basis (multi-months or-years) for climate change mitigation. The realization of this new generation of RT-AQF system will represent a significant landmark in the history of operational RT-AQF.

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